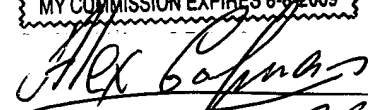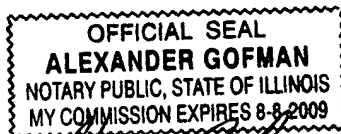## CERTIFICATE OF TRANSLATION

May 23, 2005

I, Dwight Sora, hereby certify that I am competent in both English and Japanese languages. I further certify that under penalty of perjury translation of the aforementioned patent

Patent H2000-294256

from the Japanese language into the English language is accurate and correct to the best of my knowledge and proficiency.

Professional Translator

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			Continued on last page

(54) [Title of Invention] Solid Polymer-Type Fuel Cell

(57)[Abstract of the Disclosure]
[Problem] To provide a solid polymer-type fuel cell equipped with a stainless steel separator in which the poison of each electrode-supported catalyst due to eluted metal ions is low.

[Means to Solve Problem] A solid polymer-type fuel cell equipped with a separator composed of stainless steel in which the impurities of S, P, V, Ni and CU, etc. have been lowered, and the Cr and Mo content is made to satisfy Cr: 10.5~35% and Mo: 0.5~5%, moreover, $12\% \leq Cr + 3Mo \leq 54\%$.

[Scope of Claims]

[Claim 1] A solid polymer-type fuel cell characterized by a separator being composed of ferrite stainless steel that has the chemical composition mentioned below, with a solid polymer-type fuel cell that is made to generate direct current by supplying fuel gas and oxidizing gas, on a It contains, by weight percent,

S:	Less than 0.005%,
V:	Less than 0.2%,
CU:	Less than 0.2%,
Mo:	0~6%,

and (Cr + 3Mo) is within a range of 10.5~43%

[Claim 2] A solid polymer-type fuel cell as in Claim 1 characterized by the Si and Mn in the ferrite stainless steel being, by weight percent, less than 0.3% for Si weight and less than 0.4% for Mn.

[Claim 3] A solid polymer-type fuel cell as in Claim 1 or 2 for which the C and N weight in the ferrite stainless steel is, by weight percent, C: less than 0.018%, N: less than 0.018%, and the total C and N content being less than 0.025%.

[Claim 4] A solid polymer-type fuel cell as in any of the Claims 1 thru 3 for which the ferrite stainless steel contains one or two types of Ti and Nb, Ti is less than 0.2% by weight percent and within a range of 6 (C% + N%)~25 (C% + N%), and Nb is less than 0.3% and within a range of 6C%~25C%.

[Detailed Description of Invention]

[0001]

[Field of industrial Application] This invention is related to solid polymer-type fuel cells in which a small distributed generator, such as one meant for automobile installation or one meant for domestic use, is used.

[0002]

[Prior Art] A fuel cell is a battery that generates direct current power by using hydrogen and oxygen, of which there are solid electrolyte-type fuel cells, fused carbonate-type fuel cell, phosphoric acid fuel cell, and solid polymer-type fuel cells. The name of a fuel cell is derived from the constituent material of the "electrolyte" part that basically makes up the battery.

[0003] At present, with regards to fuel cells achieving the commercial level, there are phosphoric acid-type fuel cells and fused carbonate-type fuel cells. The rough operating temperatures for fuel cells are 1000°C for solid electrolyte-type fuel cells, 650°C for fused carbonate-type fuel cells, 200°C for phosphoric acid-type fuel cells, and 80°C for solid polymer-type fuel cells.

[0004] It is easy to start and stop solid polymer-type fuel cells when the operating temperature is low at 80°C before and after, and because it is possible for the energy efficiency to be about 40%, there is anticipation for its global implementation as an emergency distributed generator for small offices and telephone exchanges, a domestic small distributed generator that assumes city gas as fuel, and a power source for installation in low-emission electric automobiles that assume hydrogen gas, methanol or gasoline as fuel.

[0005] With each of the aforementioned types of fuel cells, in cases when we think about the individual constituent laminated body that has centered a solid polymer electrolyte membrane and layered unit batteries that have superposed fuel electrode membranes and oxide electrode membranes by placing a separator between several and unit batteries.

P:	Less than 0.025%,
Ni:	Less than 0.2%,
Cr:	10.5~35%,
Rare Earth Metals:	0~0.1%

materials of items that are referred to by the common name of "fuel cell," it is necessary for them to be batched as completely different things. This is because the performance obtained by the absence of constituent materials corroding due to the electrolytes used, the absence of high-temperature oxidation that starts to actualize from around 380°C, and the absence of sublimation, reprecipitation, and coagulation of electrolytes, especially corrosion-resistance performance, is completely different for each fuel cell. Actually, the materials used are many, varying from graphite material to Ni clad material, high alloy and stainless steel.

[0006] It is not at all possible to consider the application of material used in commercialized phosphoric acid-type fuel cells and fused carbonate-type fuel cells in the constituent material of solid polymer-type fuel cells.

[0007] Fig. 1 is a drawing showing the construction of a solid polymer-type fuel cell, Fig. 1(a) is an exploded view of a fuel cell (single cell), Fig. 1 (b) is an oblique drawing of an entire fuel cell. As shown in the same drawing, the fuel cell 1 is an aggregate of single cells. A single cell is constructed, as shown in Fig. 1 (a), with fuel electrode membranes (anode) 3 layered on one face of a solid polymer electrolyte membrane 2, and oxide electrode membranes (cathode) 4 layered on the opposite face, with separators 5a and 5b superimposed on both faces.

[0008] As for a representative solid polymer electrolyte membrane 2, there is a fluorine ion exchange resin layer that has a hydrogen ion (proton) exchange group.

[0009] A catalytic layer composed of fluorine resin that has particulate platinum catalysts, graphite powder, and, as needed, a hydrogen ion exchange group, is provided on a fuel electrode membrane 3 and an oxide electrode membrane 4, and making contact with the fuel gas or oxidative gas.

[0010] Hydrogen is supplied to a fuel electrode membrane 3 by fuel gas (hydrogen or oxygen-containing gas) A being injected from a channel 6 provided on a separator 5a. In addition, from a channel 6b provided on a separator 5b, oxidative gas B such as air is injected, and oxygen is supplied. Direct current power is generated by an electrochemical reaction produced by the supply of these gases.

[0011] Anode Side: $H_2 \rightarrow 2H^+ + 2e$. Cathode Side: $(1/2) O_2 + 2H + 2e- \rightarrow H_2O$

The functions obtained by the solid polymer-type fuel cell separator are (1) function as a "channel" that uniformly supplies fuel gas in-plane on the fuel electrode side, (2)

function as a "channel" that efficiently discharges water generated on the cathode side, in addition to post-reaction air and oxygen carrier gas from the fuel cell, to outside of the system, (3) function as an electrical "connector" between the individual cells that maintains low electrical resistance and good electrical conductivity as an electrode over a long period of time, and (4) function in adjoining cells as a cathode chamber for the cell and a "partition" adjacent to the anode chamber for the cell on other side.

[0012] Up to now, the use of cathode sheet material as a separator material has come to be keenly investigated, but there has been a problem with it being "easy to fracture" the cathode sheet material, and furthermore, there have been problems with the machining cost for smoothing the surface and the machining cost for forming the gas channel becoming quite expensive. These are each unavoidable problems, and there are conditions in which they inevitably make commercialization of fuel cells difficult.

[0013] The most attention is being paid to thermally expandable graphite finished goods as material for solid polymer-type fuel cell separators because of the considerable low cost. However, in order to confer function as the aforementioned partition to reduce gas permeability, resin impregnation and baking that reaches "multiple times" must be implemented. In addition, issues to resolve in future, such as the machining costs for insuring flatness and groove formation, are many, and, at present, are not at the point that they will be realized.

[0014] With regards to activities for confronting the investigation into application of these graphite materials, attempts to apply stainless steel to the separator for the purposes of cost reduction are being started.

[0015] In the bulletin for Patent H10-228914, an announcement is made of fuel cell separators that are composed of metal material and employ gold plating directly to the face that makes contact with the electrode of the unit battery. Stainless steel, aluminum and nickel-iron alloy are provided as metal materials, while SUS304 is being used for stainless steel. In this invention, since gold plating is being employed with the separator, the contact resistance between the separator and electrode is reduced, and conduction of electrons from the separator is becomes good, so it assumed that the output voltage of the fuel cell is enlarged.

[0016] In the bulletin for Patent H08-180883, an announcement is made of solid polymer electrolyte-type fuel cells that use separators composed of a metal material on which the passivated film that forms on the surface is easily produced by air. Stainless steel and titanium alloy are provided as metal materials. In this invention, passivated film always exists on the surface of the metal used in the separator, and the amount of ionization of the water generated by an individual fuel cell by the surface

of the metal becoming chemically difficult to violate is reduced, and it is assumed that drops in the electrochemical reactivity of an individual fuel cell are inhibited.

[0017] However, even if metal material such as the stainless steel that provides the passivated film on the surface as announced in the aforementioned public bulletin is used as is on the separator, elution of metal occurs without sufficient corrosion resistance, the performance of the supported catalyst deteriorates (henceforth described as supported catalyst poison)due to eluted metal ions. In addition, because of problems with the contact resistance of the separator increased by corrosion product such as Cr-OH and Fe-OH produced after elution, at present, noble metal plating such as gold plating in which cost is disregarded is applied to separators composed of metal materials.

[0018] With regards to its use with separators for metal materials up to now, there have been results in which it has been applied, but they are at a stage far from practical implementation.

[0019] It is possible to use them as a separator with "purity" that will not have high-cost surface treatment performed, and if it is possible to develop stainless steel with excellent corrosion resistance, in addition to excellent electrical conductivity in battery environments, the manufacturing cost of fuel cells will greatly drop, and it will be possible to expect commercialization and increased use of solid polymer-type fuel cells.

[0020]

[Problems to be solved by Invention] The problem for this invention is to provide a solid electrolyte-type fuel cell equipped with a stainless steel separator in which the increase in contact electrical resistance with the electrode due to corrosion product and the increase in contact resistance due to reinforcement of the passivated film is small, without performing high-cost surface treatment, and in which the poison of each electrode-supported catalyst due to eluted metal ions is quite low.

[0021]

[Means to solve Problem] A summary of this invention is as follows below.

(1) A solid polymer-type fuel cell characterized by a separator being composed of ferrite stainless steel that has the chemical composition mentioned below, with a solid polymer-type fuel cell that is made to generate direct current by supplying fuel gas and oxidizing gas, on a laminated body that has centered a solid polymer electrolyte membrane and layered unit batteries that have superposed fuel electrode membranes and oxide electrode membranes by placing a separator between several and unit batteries.

[0023]

(2) A solid polymer-type fuel cell as in the aforementioned (1) in which the Si and Mn in the ferrite stainless steel is, by weight percent, less than 0.3% for Si weight and less than 0.4% for Mn, and which contains, by weight percent,

(4)

S: Less than 0.005%,	P: Less than 0.025%
V: Less than 0.2%	Ni: Less than 0.2%
CU: Less than 0.2%	Cr: 10.5~35%
Mo: 0~6%	Rare Earth Metals: 0~0.1%,

5

6

and (Cr+3Mo) is within a range of 10.5~43%.

[0024] A solid polymer-type fuel cell as in the aforementioned (1) or (2) for which the C and N weight in the ferrite stainless steel is, by weight percent, C: less than 0.018%, N: less than 0.018%, and the total C and N content being less than 0.025%.

[0025] (4) A solid polymer-type fuel cell as in any of the above entries 1 thru 3 for which the ferrite stainless steel contains one or two types of Ti and Nb, Ti is less than 0.2% by weight percent and within a range of 6(C%+N%)~25(C%+N%), and Nb is less than 0.3% and within a range of 6C%~25C%.

[0026] Furthermore, the separator mentioned is one that has the four functions described previously. Specifically, (a) function as a "channel" that uniformly supplies fuel gas in-plane on the fuel electrode side, (b) function as a "channel" that efficiently discharges water generated on the cathode side, in addition to post-reaction air and oxygen carrier gas from the fuel cell, to outside of the system, (c) function as an electrical "connector" between the individual cells that maintains low electrical resistance and good electrical conductivity as an electrode over a long period of time, and (d) function in adjoining cells as a cathode chamber for the cell and a "partition" adjacent to the anode chamber for the cell on other side. There are also cases of a construct to which these functions are assigned by multiple sheet layers. The separator referred to in this invention is a sheet that has at least one of the functions from among the four mentioned above.

[0027] In order to develop a solid polymer-type fuel cell equipped with a separator composed of stainless steel, the inventors implemented various tests by using single cells in the environments in which the separator is placed, for the purpose of reducing the metal ions eluted from the stainless steel surface as much as possible. According to the results, in 1~3 pH environments in which a) separators that come to complete this invention by achieving the following findings are placed (henceforth referred to simply as separator environments), the corrosion resistance of austenitic stainless steel is insufficient, and inapplicable to separators with significant elution of metal.

[0028] In separator environments b), ferrite stainless steel demonstrates good corrosion resistance, and in general ferrite stainless steel, metal elusion of a degree that will effect battery performance will be produced.

[0029] When c) metal is eluted, corrosion product (hydroxide with Fe assumed to be the subject) is generated and an increase in contact electrical resistance is brought about, causing significantly negative effects on the performance of the supported catalyst, so the battery performance represented by electromotive force deteriorates over a short time. In addition, it causes a negative effect on the positive ion conductivity of fluorine ion exchange resin layer that has a hydrogen ion (proton) exchange group.

[0030] d) In order to prevent elution of metal, the passivated film must be rigidified within the impurities inside of ferrite stainless steel, in addition to simultaneously reducing the S, P, V, Ni and Cu content.

[0031] e) Even when the passivated film is rigidified, the contact electrical resistance will intensify when the passivated film thickness is thickened, and the battery efficiency will significantly drop.

[0032] f) In order to control elution of metal in separator environments by rigidifying passivated film without thickening, it is necessary for the Cr and Mo content to be within a range of 12~43%.

[0033] g) By actively adding Mo, the corrosion resistance is insured, but even if it is assumed that the Mo has eluted, the effect on the performance of catalysts supported by anode and cathode parts is comparatively slight.

[0034]

[Mode for Invention] Below, we will give a detail explanation of the reasons for defining the chemical composition of a separator composed of ferrite stainless steel provided by a solid polymer-type fuel cell in this invention. Furthermore, the % expression of the aforementioned elements indicates weight percent.

[0035] S: intra steel S weight must be assumed to be less than 0.005%. S, corresponding to the coexisting intra steel elements and intra steel S weight, is almost completely precipitated as complex nonmetallic inclusions like Mn sulfide, Cr sulfide, Fe sulfide, Ti sulfide, and complex sulfides and oxides of these. However, in separator environments, nonmetallic inclusions of any composition also act as a source of corrosion for ones that have differences in degree, and are harmful to maintenance of passivation and control of corrosion and elution of metal.

[0036] In addition, under the conditions in which fuel cells are operated, the pH inside of the gap between separators composed of ferrite stainless steel and the MEA (Membrane Electrode Assembly) drops due to occurrence of cell reaction and/or oxygen concentration differential cell corrosion, and although conditions will arise in which micro cell corrosion easily occurs, on such occasions the nonmetallic sulfide inclusions will have large effects as sources of corrosion and acceleration factors. The intra steel S weight of regular mass-produced steel exceeds 0.005% at around 0.008%, but it must be reduced to less than 0.005% in order for the aforementioned detrimental effects to be prevented. The desirable intra steel S weight is less than 0.002%, while the most desirable intra steel S weight level is less than 0.001%, and if low, then a low level is acceptable.

[0037] P: intra steel P weight must be less than 0.025%. The content level of regular commercial stainless steel is about 0.026~0.035%. P is the inevitable impurities, and, while ranked with S, is a hazardous element that has little effect on the poisons in the anode and cathode catalytic layers. A low level is desirable.

[0038] V: intra steel V must be set to less than 0.2%. In general, V is contained as an impurity inside of the Cr source that is the essential raw dissolution material when ingoting stainless steel, while a mixture at some level is inevitable. However, the eluted V has little negative effect on the performance of catalysts supported by the anode and cathode parts. In addition to maintaining

battery characteristics, the tolerable upper limit is 0.2%, and if low, then low is acceptable.

[0039] Cu and Ni: intra steel Cu and Ni must be set to less than 0.2% for either one or both. In general, even with small impurity level amounts of Cu and Ni, there are effects of improving the corrosion resistance of stainless steel in low pH environments. However, in separator environments, even with small amounts of Cu and Ni ion elution in passivation maintenance conditions, they lead to an effect on the poisons in anode and cathode catalytic layers, so it is necessary to assume an upper limit of 0.2%. Including a mixture from scrap as well, it is more desirable for the intra steel Cu and Ni weight to be low, but Cu and Ni at an impurity level of the extremely small amount of less than 0.2% raises the passivation of ferrite stainless steel, also has the effect of controlling metal elution in a state of passivation, and, because it improves the battery performance as a result, assumes an upper limit of 0.2%.

[0040] Cr, Mo: Cr and Mo are the basic alloying elements that originally insure corrosion resistance. Corrosion resistance is as high as their content, but is a tendency towards a drop in normal temperature tenacity that accompanies Cr becoming high, and when the Cr weight exceeds 35%, mass production becomes difficult. In addition, at less than 10.5%, even if other elements are changed, it becomes difficult to insure the required corrosion resistance for the separator.

[0041] Mo is an element that is included as required, and in comparison to Cr, has the effect of improving corrosion resistance in small quantities. At less than 0.5%, the effect of Mo becomes unclear. In cases when it is included, when it is included in excess of 6%, it becomes difficult to avoid precipitation of intermetallic compounds such as Sigma phase, and production becomes difficult because of problems with steel embrittlement, so an upper limit of 5% is assumed. By actively including Mo, corrosion resistance is insured. Even when it is assumed that Mo is eluted, the effect on the performance of catalysts supported by anode and cathode parts is comparatively slight, and the negative effect on positive ion conductivity for the fluorine ion exchange resin layer that has a hydrogen ion (proton) exchange group is small.

[0042] (Cr% + 3Mo%): The stainless steel for separators is in a state of passivation in environments of 70°C to a high 100°C, which is the operating temperature for a solid polymer-type fuel cell, and requires that the contact electrical resistance be even successively low. It is necessary to control the increase in passivated film thickness and corrosion product generation within a practical range. As a required condition for this, the Cr and Mo content is the corrosion index (Cr% + 3Mo%), but it is necessary for it to be within a range of 12~43%.

[0043] It is necessary for there to be at least passivation "within 80°C aqueous sulfuric acid solution of 2.6 pH at 25°C" that is determined to be appropriate as a simulated ambient condition of the battery interior in actual operating conditions. For this to be so, it is desirable that the passivation maintenance current density be less than $50\mu A/cm^2$ at 0.2V vs. SCE in this environment.

[0044] Si: It is desirable that the intra steel Si weight be set to less than 0.3%. Si is as effective a deoxidizing element as Al in mass-produced steel. However, since eluted Si has little effect on the poisons in the anode and cathode catalytic layers, and is clearly harmful with regards to maintaining battery characteristics, less than 0.3% is assumed. Mass production at approximately 0.25% is the most desirable from the viewpoint of reducing mass production costs, but in separator environments, less than 0.2% is desirable. Even more desirable is less than 0.1%.

[0045] Mn: It is desirable that the intra steel Mn weight be assumed to be less than 0.4%. Normally, Mn has a fixing action as a Mn sulfide, and has the effect of improving hot workability. In addition, it is acceptable if it is actively added as a deoxidizing element or a Ni balance compensation element. However, even when in a state in which passivation is being maintained, metal elution barely advances at a time, but when the amount exceeds 0.4%, eluted Mn ions have little effect on the poisons in anode and cathode catalytic layers. Less than 0.1% is desirable. At less than 0.45 Mn, it is possible to manufacture without the generation of problems such as hot cracking during mass production. Hardly any problems with increased manufacturing costs will arise either.

[0046] C, N: Intra steel C and N for the purposes of improving normal temperature tenacity, C is less than 0.018%, N is less than 0.018%, with it desirable if less than 0.025% is assumed at the C%+N% value. C and N are penetrating-type elements, and become a source of deterioration in the base metal tenacity, weld corrosion resistance and tenacity, of high purity ferrite stainless steel. The strict limiting of C and N is a normal temperature tenacity measure, that becomes a problem in manufacturing processes for hot-rolled coils of high purity ferrite stainless steel, but makes it possible avoid rises in manufacturing costs. If intra steel C and N are lowered extremely, normal temperature tenacity will be improved, so if low, then a low level is acceptable.

[0047] Ti: Ti is less than 0.2% as needed, and is included by a range amount greater than six times and less than 25 times the value of (C%+N%). Ti leads to poisons in the anode and cathode catalytic layer, so it is intrinsically an element that must be reduced, but, from the viewpoint of insuring manufacturability during mass production and workability of the plating, the minimum amount is included according to need.

[0048] In cases in which weldability is not demanded, greater than (C% + N%) value x six times and less than 10 times is the most desirable. In cases in which weldability is demanded, greater than 10 times the (C% + N%) value and less than 16 times is the most desirable. In order to avoid the generation of sheet surface defects that are a source of debris, less than 0.1% is desirable. When more than needed is included, it becomes a source of anode and cathode catalytic performance degradation due to elution of metals while in a state in which passivation is maintained.

[0049] Nb: Nb is an element that is included as needed, and is also an alloying element for which the bonding strength with intra steel C and N is stronger than Cr, the same as Ti. Nb is less than 0.3%, and is included within a range of C% x 6~C% x 25 [specifically, Nb (%) / C (%) = 6~25%]. It is quite effective in improving tenacity, including normal temperature tenacity of hot-rolled coils. However, Nb eluted along with corrosion accumulates as a corrosion product on the corrosion face, and has the harmful effect of raising contact electrical resistance, so it is more desirable if the Nb content is low from the viewpoint of base material performance. However, the required minimum amount is added in the cases of insuring weld performance, or determining the necessity of improving the workability of cold-rolled steel sheet material by simultaneously including Nb and Ti.

[0050] Rare Earth Metals (REM): The bonding strength with S at the molten steel stage is quite strong with rare earth metals, so it has the effect of making S harmless. Accordingly, it is acceptable if it is added in a form like misch metal as needed. Content achieves sufficient effects at less than 0.1%.

[0051] It is acceptable if elements other than the aforementioned elements are included as needed. For example, it is good to include Ca, Mg or B less than 0.1 to improve hot workability.

[0052]

[Working Example] We dissolved ferrite stainless steel of the 28 types of chemical compositions as shown in Table 1 in a high frequency induction heating method 150 kg vacuum melting furnace. We carefully selected and used material with few market impurities as the raw material for melting, and the adjusted the amount of intra steel impurities.

[0053]

[Table 1]

表1 (Weight Percent, Residual Fe)

		11 Example of this Invention														12 Comparative Example		

No.	C	Si	Mn	P	S	Cu	Ni	Cr	Mo	Ti	V	N	Al	Other	C+N	6(C+N)	25(C+N)	Cr-3Mo
1	0.0025	0.08	0.08	0.012	0.0008	0.006	0.003	17.6	0.82	0.10	0.007	0.005	0.012		0.0075	0.0450	0.1875	20.06
2	0.0018	0.12	0.08	0.010	0.0008	0.008	0.004	18.3	0.82	0.10	0.008	0.005	0.010		0.0068	0.0408	0.1700	20.76
3	0.0019	0.15	0.06	0.011	0.0006	0.005	0.003	19.5	1.20	0.11	0.006	0.005	0.008		0.0069	0.0414	0.1725	23.10
4	0.0022	0.10	0.09	0.012	0.0009	0.005	0.005	20.5	1.81	0.10	0.003	0.006	0.011		0.0082	0.0492	0.2050	25.93
5	0.0013	0.08	0.10	0.011	0.0008	0.008	0.004	25.5	0.98	0.12	0.007	0.005	0.010		0.0063	0.0378	0.1575	26.54
6	0.0020	0.08	0.08	0.012	0.0009	0.005	0.005	19.5	0.52	0.11	0.006	0.004	0.011		0.0060	0.0360	0.1500	20.06
7	0.0018	0.10	0.08	0.010	0.0006	0.005	0.005	19.9	1.82	0.11	0.008	0.006	0.012		0.0078	0.0468	0.1950	24.60
8	0.0013	0.09	0.06	0.011	0.0007	0.005	0.004	19.5	2.82	0.11	0.007	0.005	0.010		0.0066	0.0396	0.1650	27.96
9	0.0012	0.11	0.10	0.012	0.0003	0.006	0.003	29.2	4.02	0.11	0.005	0.005	0.013	La:0.08	0.0062	0.0372	0.1550	41.26
10	0.0016	0.10	0.09	0.010	0.0008	0.013	0.003	19.8	1.88	0.10	0.003	0.005	0.012		0.0076	0.0456	0.1900	24.84
11	0.0025	0.08	0.10	0.009	0.0009	0.012	0.012	19.2	2.02	0.10	0.006	0.006	0.010		0.0085	0.0510	0.2125	25.26
12	0.0015	0.08	0.06	0.007	0.0007	0.012	0.015	19.3	0.82		0.005	0.005	0.011		0.0066	0.0396	0.1650	21.46
13	0.0015	0.06	0.08	0.006	0.0003	0.011	0.015	19.3	1.95		0.008	0.005	0.012		0.0065	0.0390	0.1625	25.15
14	0.0015	0.08	0.08	0.006	0.0006	0.013	0.016	18.3	2.86		0.008	0.005	0.013		0.0065	0.0390	0.1625	27.48
15	0.0022	0.32	0.43	0.008	0.0005	0.010	0.012	22.3	1.82		0.003	0.003	0.010		0.0052	0.0312	0.1300	27.76
16	0.0025	0.08	0.08	0.007	0.0070*	0.010	0.011	16.8			0.120	0.008	0.010		0.0105	0.0630	0.2625	16.80
17	0.0022	0.28	0.09	0.007	0.0075*	0.010	0.008	16.5			0.120	0.007	0.009		0.0092	0.0552	0.2300	16.50
18	0.0020	0.06	0.45	0.008	0.0065*	0.009	0.009	16.8			0.110	0.007	0.010		0.0090	0.0540	0.2250	16.80
19	0.0016	0.06	0.45	0.029*	0.0068*	0.150	0.010	16.8			0.130	0.006	0.008		0.0076	0.0456	0.1900	16.80
20	0.0015	0.30	0.07	0.028*	0.0005*	0.750*	0.120	16.7			0.110	0.007	0.010		0.0085	0.0510	0.2125	16.70
21	0.0038	0.05	0.08	0.007	0.0058*	—	—	19.2			0.110	0.005	0.008	Nb:0.48	0.0088	0.0528	0.2200	19.20
22	0.0041	0.08	0.08	0.008	0.0065*	—	0.152	19.0			0.120	0.006	0.009		0.0101	0.0606	0.2525	19.0
23	0.0200	0.05	0.06	0.006	0.0072*	0.120	—	19.2	1.80		0.110	0.009	0.018		0.0290	0.1740	0.7250	24.6
24	0.0250	0.45	0.50	0.028*	0.0085*	0.120	0.080	11.5	0.02		0.250*	0.020	0.002		0.0450	0.2700	0.1250	11.56
25	0.0280	0.25	0.45	0.030*	0.0075*	0.230*	0.100	18.5	0.02		0.080	0.023	0.003		0.0510	0.3080	1.2750	16.86
26	0.0580	0.34	1.20	0.030*	0.0061*	0.240*	8.210*	18.5	0.03		0.070	0.031	0.002		0.0890	0.5340	2.2250	18.74
27	0.0130	0.62	1.00	0.028*	0.0062*	—	12.30*	17.5	2.24			0.023	0.003		0.036	0.2160	0.9000	24.22
28	〃	〃	〃	〃	〃	〃	〃	〃	〃		〃	〃	〃		〃	〃	〃	24.22
29	0.0230	0.24	0.53	0.025	0.0230	0.080	0.020	10.1	—		0.030	0.005	0.002		0.028	0.168	0.7000	10.10

*Indicates outside of the range established by this invention.

[0054] Ingots in which the ingot-casted cross-section is round, after being heated for three hours at 1230°C in air, are hot forged by a press method forging machine, and then each ingot is finished on a test slab of the following two types.

[0055] ① Thickness 30 mm, Width 100 mm, Length 120 mm
② Thickness 70 mm, Width 380 mm, Length 550 mm

The slab in ① is assumed to be hot-rolled into a steel sheet of 6 mm thickness, and then cooled while wrapped in heat insulating material that simulates temperature history immediately after the completion of hot-rolling in mass production. Then it is supplied to a Charpy impact test in order to find the tenacity of the hot-rolled coil at room temperature. The specimen is assumed to be a JIS Z2202 No. 4 half size.

With the slab in ②, the slab surface is cut by machine, the oxided scale of the surface is removed, and then finished on a slab of 62 mm thickness. This slab is heated to 1200°C in air, and after being finished to a 4 mm thickness by hot rolling, it is annealed while wrapped in heat insulating material that simulates temperature history

① No. 25 hot-rolled steel sheet in Table 1 .... 830°C
② No. 11~15, 16~23 hot-rolled steel sheet in Table 1 .... .900°C
③ No. 1~10 hot-rolled steel sheet in Table 1 .... .930°C
④ No. 24 and 29 hot-rolled steel sheets in Table 1 .... .980°C
⑤ No. 26 and 28 hot-rolled steel sheets in Table 1 .... 1080°C

At each temperature, re-crystallization progressed, and the temperature at which intermetallic compounds dissolve was assumed. In-furnace time was about 20 minutes.

[0057] Next, the solution annealed hot-rolled steel sheet was finished at a 0.3 mm thickness without cold rolling, while using a multistage Sendzimir-type rolling mill and holding the intermediate annealing midway. The final finishing annealing is performed in a bright annealing furnace for a hydrogen atmosphere in which the dew point is less than -50°C, with the temperature assumed to be the same as the annealing temperature for hot-rolled material. The holding time was one minute, while the in-furnace time was approximately three minutes.

[0058] From this cold-rolled annealing material, the loading separator for specimens meant for performing evaluations of the passivated film in a simulated ambient separator conditions of the following dimensions, and actual solid polymer-type fuel cells, is manufactured from a press form.

[0059] Furthermore, with comparative example No. 27, after manufacturing samples for simulated ambient conditions and separators, we performed one-sided gold plating of thickness 5 μm.

[0060] Specimens for Simulated Ambient Conditions:
Thickness 0.3 mm, Width 10 mm, Length 10 mm
Separator:
Thickness 0.3 mm, Vertical 80 mm, Horizontal 80 mm
Gas Channel: Height 0.8 mm, Peak-to-Peak Gap 1.2 mm

immediately after the completion of hot-rolling in mass production, the same as in ①.

[0056] We performed solution annealing maintained for four minutes at the following temperatures corresponding to the chemical composition, and then forced air cooled.

(Corrugated Processing) These surfaces were mechanically shot ground finished by using SiC abrasive for shot working, ultrasonically cleaned for 15 minutes at 5%HNO₃ + 3%HG and 40°C, alkali spray degreased immediately before testing by using 6% aqueous sodium hydroxide solution, and then, after a simple cleaning with water, soak washed three times with distilled water in a batch-type water tank, and then after further cleaning with distilled water spray for four minutes and drying with a cold blast dryer, were supplied to each test.

[0061] In the simulated ambient conditions tests, the temperature of 2.6 pH aqueous sulfuric acid solution was raised to 80°C when at a temperature of 25°C reached by using guaranteed reagent sulfuric acid, samples were soaked for six hours in this solution, and then, in addition to evaluating the presence of passivation from the mass loss, the hydrogen bubble generation from the material surface and discoloration of the test solution, in order to more carefully examine the level of metal elution, we measured the passivation maintenance current density at 0.2V vs. SCE.

[0062] Charpy impact test results and the results of tests in simulated ambient separator conditions were as shown in Table 2.
[0063]
[Table 2]

No.	Evaluation in 2.6 pH Aqueous Sulfuric Acid Solution at 25°C		Evaluation according to Loading of a Single Solid Electrolyte-Type Fuel Cell	Impact Value at the Normal Temperature (25°C) for a Hot-Rolled Steel Sheet Air-Cooled after Pressing	Notes
	Presence of Passivation	Passivation Maintenance Current Density at 0.2V vs. SCE ($\mu A/cm^2$)	Rate of Voltage Decrease for a Single Cell after One Hour Elapse 1-{Voltage V after Elapse/Initial Voltage v}	(Specimen: JIS-4 Half Size (kgf-m/cm²)	
1	Yes	10~20	<0.05	>15	
2	"	20~30	<0.05	>15	
3	"	10~20	<0.05	>15	
4	"	10~20	<0.05	>15	Example of this Invention
5	"	10~20	<0.05	>15	
6	"	10~20	<0.05	>15	
7	"	<10	<0.05	>15	
8	"	<10	<0.05	>15	
9	"	<10	<0.05	>15	
10	"	10~20	<0.05	>15	
11	"	10~20	<0.05	>15	
12	"	<10	<0.05	>15	
13	"	<10	<0.05	>15	
14	"	<10	<0.05	>15	
15	"	<10	<0.05	>15	
16	No	50~70	0.3~50	2	
17	"	50~70	0.3~0.5	2	
18	"	50~70	0.3~0.5	3	
19	"	50~70	0.3~0.5	2	
20	"	50~70	0.3~0.8	2	Comparative Example
21	Yes	50~70	0.3~0.5	3	
22	"	50~70	0.3~0.5	3	
23	"	50~70	0.5~0.7	1	
24	"	>1mA/cm²	>0.8	12	
25	"	50~70	>0.8	12	
26	"	10~20	>0.8	25	
27	"	10~20	>0.8	28	
28	"	<10	<0.05	28	
29	"	>1mA/cm50~70	>0.8	18	

[0064] As clearly shown in Table 2, this invention is in a state of passivation within a 2.6 sulfuric acid solution at a temperature of 80°C up from 25°C, with the "passivation maintenance current density" that indicates the level of elution also less than 20 $\mu A/cm^2$.

[0065] It has not been said that it is desirable to assume a passivation maintenance current density at as low a level as possible, when stainless steel is used as a separator for a solid polymer-type fuel cell. It is important to be stable and achieve a low level, and although less than 10 $\mu A/cm^2$ is the most desirable, the next desirable is 10~20 $\mu A/cm^2$.

[0066] The passivation current density in the comparative example is 30~80 $\mu A/cm^2$, and although it can be said to be controlled, it indicates that it is in a state in which comparatively large elution from the separator is taking place.

[0067] The inventors determined that performance is insufficient at passivation maintenance current density > 50 $\mu A/cm^2$ from the relationship with the performance characteristics of an actual single cell battery as the standard for determining the applicability of a fuel cell separator material. With materials for which passivation maintenance current density is less than 50 $\mu A/cm^2$, even evaluation tests for a single actual cell have not been confirming successive performance deterioration at a level that is a problem, and have been determining the high appropriateness of this as an evaluation condition for rapid

simulated ambient conditions. Even in these results, the case of the invention example is at the same level as gold plated materials that are one of the most desirable metal materials in solid polymer-type fuel cell environments (sample steel 27), and it has been determined that their relatively good performance can be insured.

[0068] Next, we evaluated the characteristics while in a loaded state as a separator inside of an actual solid polymer-type single cell battery, measured the voltage of the single cell battery after one hour elapsed from injection of fuel gas into the battery interior, and then examined the rate of voltage decrease according to comparison with the initial voltage. Furthermore, the rate of decrease was obtained according to 1 – (Voltage V after One Hour Elapse/Initial Voltage v).

[0069] The solid polymer-type single fuel cell battery used for evaluation was a reworked commercial battery cell FC50 made by ElectroChem, Inc. of the U.S.A.

[0070] We used 99.9999% hydrogen gas as the anode electrode side fuel gas, and air as the cathode electrode side gas. In addition to the battery body heating the entire body to 7 8± 2°C, it adjusted the humidity control of the battery interior on the side that the exhaust gas water concentration measurement for the cell output side was originally placed. The pressure of the battery interior is 1 atmosphere. The pressure of gas introduced into the battery such as hydrogen gas and air was adjusted at

0.4~0.20 bar. For cell performance evaluation, successive measurements were performed according to conditions in which it was possible to confirm 500 mA/cm^2-0.62V at single cell voltage.

[0071] As for the single cell performance measurement system, we reworked and used a fuel cell measurement system that assumed the 890 Series made by Scribner of the U.S.A. as the base. It is possible to predict changes in characteristics according to battery operating conditions, but this is a comparative evaluation under the same conditions.

[0072] Results are shown in Table 2.

[0073] As clearly shown in Table 2, in this invention, the rate of voltage decrease is overall less than 0.05, and becomes the same as with a separator with high corrosion-resistant gold plating at a No. 27 high cost. In addition, in the comparative example that deviates from the chemical composition established in this invention, the rate of voltage drop was quite large at 0.3~0.8.

[0074] In one part of the specimens, an evaluation using long-term tests was implemented, but results that approximately correlated with the short-term test results shown in Table 2 were achieved.

[0075] With regards to the tenacity of stainless steel at normal temperature, ferrite stainless steel is generally inferior in comparison to austenitic stainless steel. However, as clearly shown by the Table 2 Charpy test results, the intra steel C and N content is high, and, in comparison to the case of the comparative example, the case of the series of invention examples with low C and N content has considerably excellent tenacity. In general, normal temperature tenacity will apparently improve when the sheet thickness thins, so if it is at the level in the case of this invention, there will be absolutely no problems in practical use. Specifically, it can be said that the normal temperature tenacity of a hot-rolled coil that becomes a problem when manufacturing high purity ferrite stainless steel is good. In general, normal temperature tenacity will apparently improve when the sheet thickness thins, so if it is at the level of the steel in this invention, there will be absolutely no problems in practical use.

[0076]

[Effects of Invention] A solid polymer-type fuel cell of this invention has a separator that is composed of ferrite stainless steel, and does not require high-cost gold plating, and so it can be manufactured at low cost.

[Brief Description of Drawings]

[Fig. 1] Drawing showing the construction of a solid polymer-type fuel cell.

[Explanation of Numbers in Drawing]

1	Fuel Cell
2	Solid Polymer Electrolyte Membrane
3	Fuel Electrode Membrane
4	Oxide Electrode Membrane
5a, 5b	Separator

[Fig. 1]
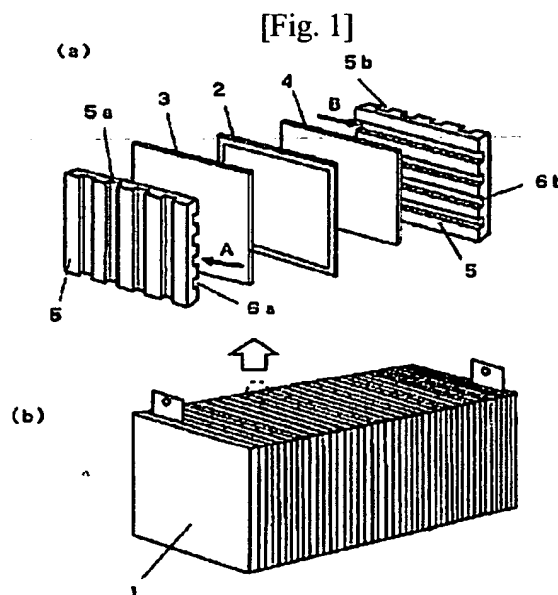
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